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CdSe quantum islands in ZnSe: a new approach

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Abstract

While providing a general overview over the current status of self-organization of quantum islands in the II–VI semiconductor system, with the main focus on CdSe embedded in ZnSe, this paper shall give an introduction to the possibilities opened by a modification of the standard growth technique. In molecular beam epitaxy, we have substituted the generally used Cd-elemental source with a CdS-compound source. The sulfur is usually not included in the growing layer. However, its presence can be surfactant-like while the elevated Cd-temperature of the dissociated CdS leads to changed thermodynamic conditions on the growth front. Using migration enhanced epitaxy, nearly perfect quantum wells with respect to lateral homogeneity can be obtained by suppressing the inherent Cd segregation and clustering. These processes are generally responsible for the formation of small islands (SI) (lateral diameter 3–5 nm) even when not attempting to grow island like structures. The suppression of these SI was a first step to gain control over the island formation. Larger islands with central Cd concentrations above 40% are more of interest for device applications, since a population at room temperature is necessary. In particular, high-density systems are required. Using the modified growth mode, well correlated, stacked island systems were obtained. Their outstanding structural and optical properties will be discussed in detail. The absence of a closed wetting layer in the CdSe/ZnSe system and the appearance of island like structures, even at submonolayer nominal deposition, further corroborate the assumption that island formation does not readily occur in a standard Stranski–Krastanow growth mode, which is assumed for InAs/GaAs. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

The large variety of new possible applications exploiting the quasi zero-dimensional nature of quantum dots (QD) has drawn attention to self-organizing semiconductor systems, such as the well investigated InAs/GaAs [1] and II–VIs like CdSe/ZnSe [2–4] and CdTe/ZnTe [5,6]. The latter system would be suitable for optoelectronic devices in the green visible spectral range. InAs QDs form on GaAs in the Stranski–Krastanow (SK) mode [7]. Elaborated techniques like indium flush [8] allow for very controlled embedding and coherent

stacking of the QDs. With this method, the first outstanding near infra-red emitting laser devices have been demonstrated by various groups [9]. For CdSe-based islands, the formation mechanisms are not completely understood, and recent results indicate that the formation does not occur in an SK mode, but rather is driven by a strong tendency of segregated Cd to form clusters. The strong differences in growth kinetics and island formation mechanisms are responsible for the very different behavior of CdSe based islands, as compared to the classical InAs/GaAs-system. In particular, to obtain a controlled island formation with predictable island sizes and ordering appears to be difficult as the competition between island formation and misfit dislocation formation as strain relaxing mechanisms plays an important role. Furthermore, a surface activated interdiffusion occurs between Cd and Zn which leads to washed out and broadened Cd concentration profiles

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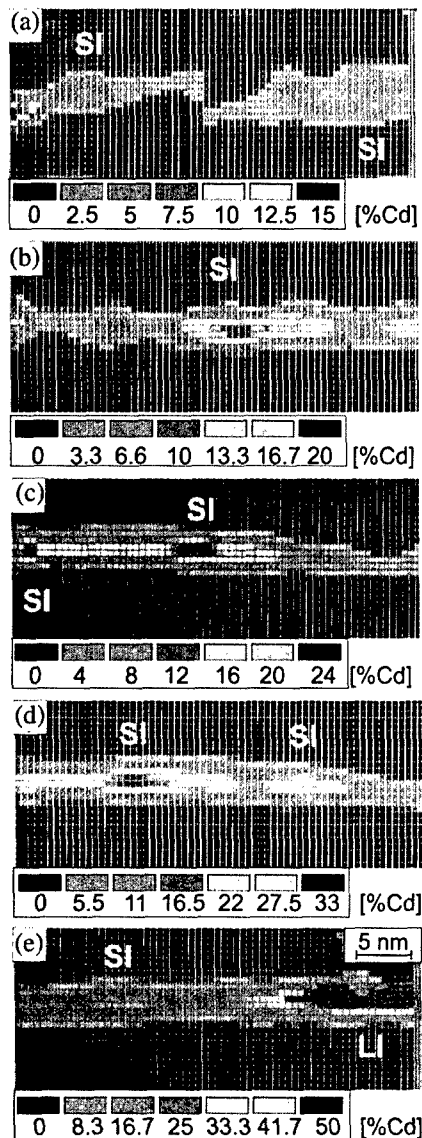


Fig. 1. Color coded Cd concentrations obtained by CELFA for a series of CdSe/ZnSe layers with varying nominal CdSe deposition: (a) 0.5 ML; (b) 1 ML; (c) 1.5 ML; (d) 2.5 ML; and (e) 3.5 ML. Cd concentrations are given in [%] according to the color code. Cd accumulations corresponding to SIs are observed even for 0.5 ML; one LI was captured for 3.5 ML.

[10,11]. In the following, we will discuss the implications and demonstrate ways to partially overcome some of these problems.

2. Classical growth methods

Using molecular beam epitaxy (MBE) under standard growth conditions (employing elemental Cd and Se at growth temperatures in the range of 270–320 °C), island formation has been demonstrated by various groups [2,12,13]. First indications that quasi-zero-dimensional states might play a role in the strong lateral localization observed in CdSe/ZnSe submonolayer structures date

back to 1993 [14]. Nowadays, it is common knowledge [15–18] that a type of smaller island is generally formed when embedding CdSe in ZnSe, which we will refer to in the following as ‘small islands’ (SI) for simplicity. Fig. 1 demonstrates this unintentional type of islands which manifests in Cd accumulations of typically 3–5 nm lateral diameter in a series of quantum wells (QW). Composition evaluation by lattice fringe analysis (CELFA) [19] allows a direct access of the Cd concentration in ternary (Cd, Zn)Se alloys with respect to pure ZnSe. The color code is directly related to the Cd concentration as given in the individual figures. The thickness of the QWs was varied from 0.5 to 3.5 ML deposited by controlled atomic layer epitaxy (ALE) where one shutter cycle corresponds to approximately 0.5 ML [20]. Surface activated interdiffusion leads to the generally broad Cd distribution in growth direction. A strong indication for this mechanism is the fact that all distributions, shown in Fig. 2, have basically the same width (≈ 11 ML), while the central Cd concentration increases with increasing nominal deposition and is independent of the total cap layer thickness. The appearance of islands even for a sub-monolayer deposition and the absence of a closed wetting layer corroborate the assumption that SIs do not form in an SK mode.

The SIs are of general interest for investigating fundamental physical properties of quasi-zero-dimensional systems [5,21–24], in particular, lateral interactions, which can be observed by time and temperature resolved optical measurements. The SI, however, are not suitable for room temperature device applications since they are unoccupied due to thermal activation at temperatures above 150 K [25,26]. The time-evolution of the photoluminescence (PL) for the 2.5 and 3.5 ML QWs containing SIs from Fig. 1 is shown in Fig. 3. A strong lateral interaction is indicated by a pronounced

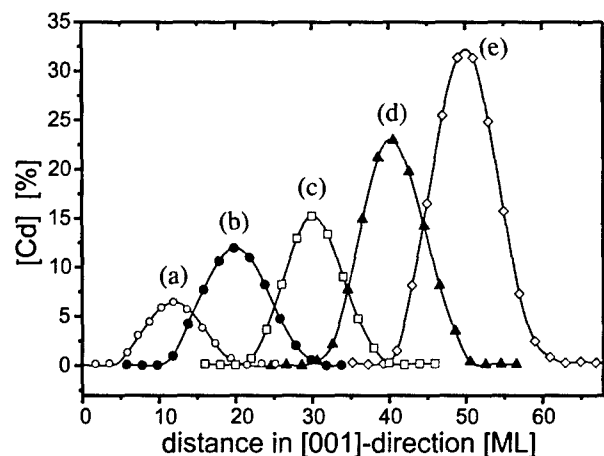


Fig. 2. Cd concentration profiles extracted from Fig. 1 with a typical, approximately Gaussian distribution. The labels correspond to (a–e) of Fig. 1; the curves are shifted horizontally for clarity.

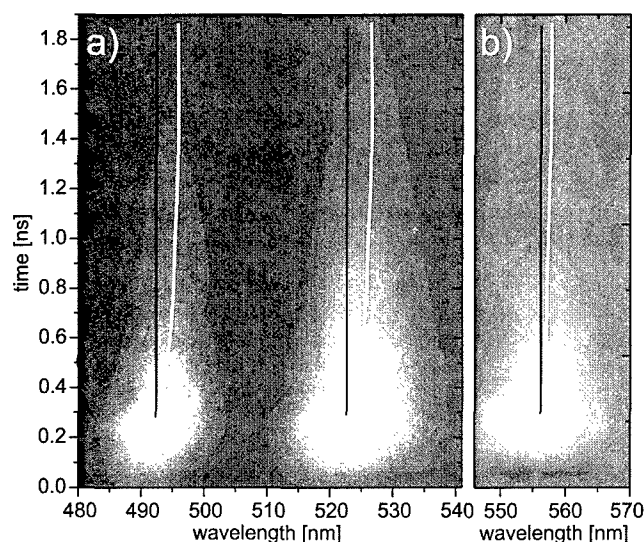


Fig. 3. Time resolved PL maps for: (a) the two thicker QWs of Fig. 1 consisting of laterally strongly interacting SIs. (b) A sample containing LIs with weak lateral interaction, mirroring in a reduced red shift with time. The evolution of the respective PL maxima has been indicated by a white line.

red shift of the luminescence emission peak, which is explained by the lateral transfer of excitons into deeper neighboring islands with time. This is only possible because the typical SI densities are of the order of $5 \times 10^{11} - 10^{12} \text{ cm}^{-2}$. The appearance of phonon replica with a distinct temporal evolution fingerprint points to a recombination of the transfer exciton in a different island and gives a hint of how this transfer is achieved: by phonon assisted hopping [27].

Large islands (LI) with a higher central Cd concentration of more than 40% and diameters of the order of 10 nm are desired for potentials deep enough to allow a room temperature population [18]. Such islands are formed if the nominal CdSe coverage exceeds ≈ 1.8 ML [28]. Density and sizes can roughly be controlled by intentional Ostwald ripening, i.e. introducing short growth interrupts of the order of few seconds. LI densities are typically of the order of $1 - 5 \times 10^{10} \text{ cm}^{-2}$ and are possibly more comparable to the SK islands observed in the III–V system InAs/GaAs. The lateral interaction between these islands is weaker due to the larger average distance, and the pronounced red shift observed in Fig. 3b is reduced in comparison with the SI, since excitons recombine in the LI in which they were captured initially; see also [13,29].

It has to be noted that the formation mechanism of the LIs is not yet understood. These islands are subject to a strong Ostwald ripening when left uncapped if the nominal CdSe deposition exceeds 2.5 ML [28] and can be associated with a stacking fault when exceeding a maximum size [30–32].

In order to control the formation of LIs, techniques such as low temperature ALE have been introduced [3,13,33,34]. However, it would be desirable to avoid the extensive temperature ramps when growing multi-layer stacks. Introducing sulfur in the growth process by using CdS as a Cd supply seems to allow a rather reproducible formation of CdSe-based LIs. The islands are directly formed at the standard growth temperature of the ZnSe buffer layer at 280 °C.

3. Introducing sulfur by using a CdS compound instead of elemental Cd in MBE

In order to gain control over the island formation, we first tried to suppress island formation all together. The technique that proved to be most suitable was to substitute the commonly used Cd-elemental source with a CdS-compound source and use a migration enhanced growth mode with long waiting times after the Cd deposition to allow excess Cd to chemically bind onto the surface [11]. While the interface driven interdiffusion remains a problem (as seen in Fig. 4), the Cd segregation and clustering could be almost completely suppressed, as demonstrated by the rather sharp interfaces and the plateau-like Cd distribution observed for these QWs. Few Cd accumulations are observed which may be associated with stacking faults propagating and penetrating the layer from the GaAs/ZnSe interface. The strain field leads to an attraction of Cd or even an enhanced interdiffusion Cd/Zn, which is consistent with the findings Shubina et al. [35] and Lürßen et al. [36]. The photoluminescence (PL) emission spectral half-width (shown in [11]) is, at 12 and 16 meV, respectively, close to the theoretical limit for ternary QWs, where a random distribution of the Cd leads to an inhomogeneous broadening [37,38]. Calculations according to the model of Mathieu et al. [39] lead to the suggestion that in the calculations used, the valence band offset between CdSe/ZnSe of 16% [40] needs to be corrected to higher values when considering the absence of SIs, since this type of laterally varying potential was neglected in the early measurements of (Cd,Zn)Se-based quantum structures [11].

Using a CdS compound source has several advantages. The oven temperature is, at 650 °C, much higher than for elemental Cd (220 °C) allowing the incoming atoms/molecules to transfer a higher thermal energy to the surface, which possibly leads to a higher surface temperature and a higher mobility or diffusion length of adatoms. As has been shown by Goldfinger et al. [41], CdS decomposes into atomic Cd and sulfur dimers when desorbed. The low sticking coefficient of sulfur with respect to Se, therefore, leads to only a minor sulfur contamination $\leq 2\%$ [42]. Schikora et al. [43] have recently shown by the observation of the recovery of a streaky 2D-RHEED (reflection high energy electron

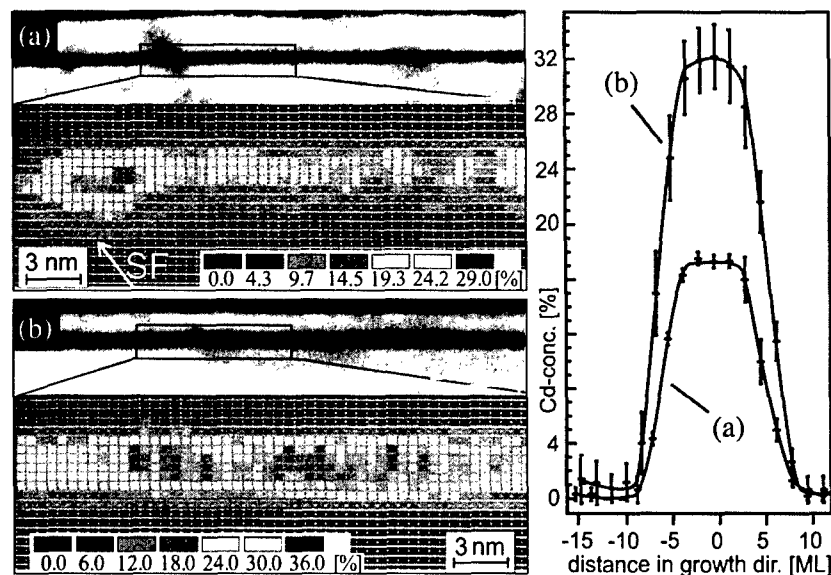


Fig. 4. CELFA of two quantum wells grown by CdS + Se MEE. Nominal CdSe deposition: (a) 1.6 ML; (b) 3.2 ML. Panel (c) shows the Cd concentration profiles extracted from (a) and (b). The steepness of the flanks is mainly resolution limited.

diffraction) pattern, typical for flat surfaces, that the presence of a massive sulfur flux may cause a resolution of previously formed CdSe islands. In our case, only a small amount of sulfur is present at the surface, which may act as a surfactant improving the CdSe nucleation. In case of the QWs, the long waiting times lead to a complete desorption of the sulfur, while in the case of continuous growth the presence of the sulfur seems to enhance the nucleation of islands. In that matter, a more controlled island formation was achieved, which enabled us to coherently stack CdSe LIs.

4. Stacked CdSe islands

To truly exploit the strong enhancement of the oscillator strength in a narrow spectral range expected for a quasi-zero-dimensional QI system, it is necessary to guarantee a narrow ensemble size distribution and to enhance the island density. This is best achieved by subsequently stacking multiple QI layers with a suitable spacer thickness. In the III–V case, elaborate techniques allow for very good control of island stacks, which have been used in the first devices [44].

Very few reports exist for II–VI systems of correlations in stacked layers [6,31,32,45]. Early reports [45] cover the dependence of correlation and anti-correlation in short period submonolayer CdSe/ZnSe stacks on stacking conditions such as barrier thickness. Litvinov et al. [31,32] demonstrated coherently stacked islands obtained at elevated growth temperatures of 400 °C in triple stacks if the barrier thickness corresponded to 12–20 monolayers (ML), while for a smaller thickness, a complete intermixing due to interdiffusion occurred. To study stacked LIs, we have chosen a spacer layer

thickness of 4 nm, i.e. 14 ML. Even though the formation of LIs may not occur in the SK mode, the strain induced by an underlying layer of islands leads to a favorable accumulation of Cd on top of these islands. Stacks with very similar appearance, as in comparable InAs/GaAs structures, are formed, which can be seen in Fig. 5. In this example, an unusually high central Cd concentration above 80% has been observed; for further details, see Kurtz et al. [16]. The resulting contrast inversion allows the island stacks to be easily distinguished from the surrounding regions, which consist of laterally strongly interacting SIs [25,27]. This region will be referred to as uncorrelated region (UR) in the following, since no direct positional correlation could be observed for SIs in subsequent layers in CELFA. In PL, two transitions can be distinguished, which are assigned to the emission from the correlated LIs and the UR, as shown in Figs. 6 and 7. While similar spectra are observed from InAs/GaAs single layers showing wetting layer and island emission, in the case of CdSe/ZnSe based single layer structures, only one emission band is observed in general. This emission band corresponds to the LI band if a sufficient number of radiating LIs are present or to the SI band if the LI density is too low. The stacking appears to improve the ordered LI formation. The high quality and homogeneity of the stacks is demonstrated by the rather narrow PL linewidth of typically 30–40 meV. It has to be noted, however, that initially formed islands are partially dissolved in the cap layer [42], which changes the appearance of the islands to a pancake or lens shape. The emission energy is more two-dimensional like, and seems to be determined by the island height rather than the lateral size, which generally exceeds the exciton

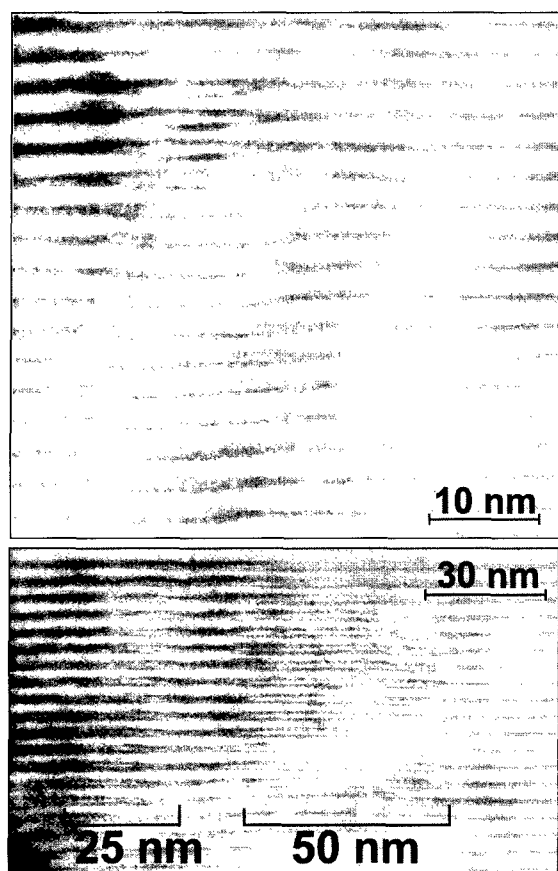


Fig. 5. Cross-sectional (002) dark field TEM images of a sample with 50 stacked layers with 4 nm ZnSe spacers. The island stacks are clearly observable by the contrast inversion owing to a rather high central Cd concentration of more than 80% [16].

Bohr radius and can vary between 20 and 50 nm within one sample. Nevertheless, the lateral size remains roughly constant within a single stack, as seen in Fig. 5.

With an increasing number of stacked layers (2, 4 and 8) the UR-PL band is enhanced as shown in Fig. 6. Two reasons could be responsible. The higher probability of finding deeper localization sites within the SI contained in the UR by vertical interaction can reduce the transfer of excitons from the UR to the LI stack. Even more reasonable appears to be the saturation of the LI emission due to the strong vertical interaction of the stacked islands. A vertical localization is expected within one stack due to variations in the island sizes. Thus, the stack states can be filled and the number of deepest LI states does not increase with increasing the number of stacked layers, while the weaker vertical interaction of the SIs leads to an increasing number of SI states proportional to the number of stacks, which can be seen in an approximate doubling of the integrated intensity of the UR peak for twice the number of layers; see Fig. 6. Furthermore, only a minor red shift occurs for the UR region when increasing the number of layers,

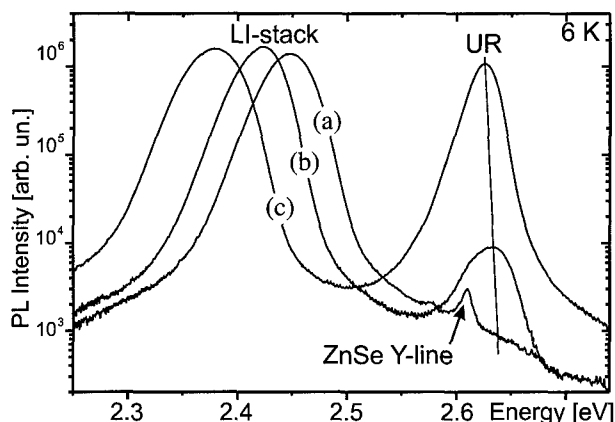


Fig. 6. Photoluminescence spectra for a series of stacked CdSe layers with constant nominal CdSe deposition (≈ 2 ML) and varying number of layers: (a) two; (b) four; and (c) eight layers, respectively. The barrier width was set to 4 nm.

showing that these layers are mainly independent while the strong red shift of the LI stack corroborates the assumption of a strong interaction within a single stack.

As shown in Fig. 7, an increasing total CdSe deposition at a fixed number of stacked layers of 12 leads to an enhanced splitting of the LI and UR emission, which is an indication of the increasing height of the LI islands with respect to the SI, while both types gain in the central Cd concentration.

Looking at the temperature dependent PL intensity of the intermediate sample (b) of Fig. 7, plotted in Fig. 8, a strong transfer of excitons into the LI stack is observed by the enhancement of the LI emission at temperatures above 50 K. Excitons are easily thermally activated

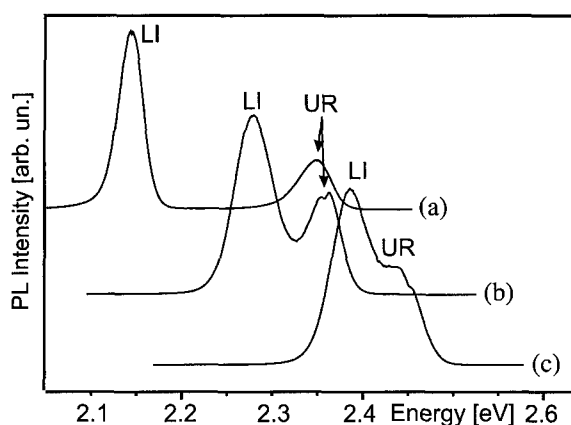


Fig. 7. Photoluminescence spectra for a series of stacked CdSe layers with constant number of layers (12) and varying nominal CdSe deposition: (a) 3–4 ML; (b) ≈ 2.5 ML; and (c) ≈ 2 ML. The CdSe growth rate was approximately twice as fast than for the previous series leading to a smaller LI–UR separation, as smaller islands are formed. Sample (a) contains stacking faults bounded by 60° -dislocations starting in the first CdSe layer. The islands are aligned along these similar to [35].

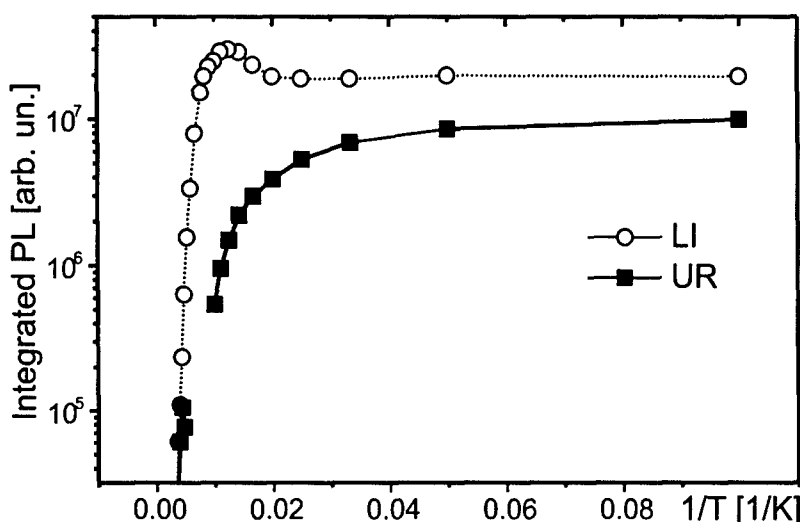


Fig. 8. Arrhenius plot of the PL intensity vs. the inverse of temperature demonstrating the thermal activation of excitons from the UR (full squares) which are then captured by the LI (open circles), indicated by a strong increase of the LI intensity at temperatures above 50 K.

from their localization in the SIs into the continuum of the UR as mentioned in Section 2. These excitons can then be recaptured by the LI stack, thus feeding the LI emission with an activation energy of $\approx 8\text{--}9$ meV corresponding to 90–100 K. The activation energy for a depopulation of the LI, at ≈ 70 meV, corresponds reasonably well to the energy separation of 82 meV between LI and UR emission peaks observed at low temperatures.

5. Conclusions

Using a modified growth technique we have demonstrated that the suppression of Cd/Zn segregation can lead to QWs with very weak lateral confinement. In contrast, a better control over the island formation is achieved and it was possible to obtain well-correlated CdSe-based quantum island stacks. The optical properties gave a first insight into interactions between the various types of islands and will allow a very detailed study by time- and spatially resolved PL in the future to gain a better insight into the fundamental physical transfer mechanisms and types of interactions.

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